

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
11 July 2002 (11.07.2002)

PCT

(10) International Publication Number  
**WO 02/053824 A1**

(51) International Patent Classification<sup>7</sup>: **D06L 1/04**

(21) International Application Number: **PCT/EP01/15048**

(22) International Filing Date:  
19 December 2001 (19.12.2001)

(25) Filing Language: **English**

(26) Publication Language: **English**

(30) Priority Data:  
00311784.3 29 December 2000 (29.12.2000) EP

(71) Applicant (for all designated States except AG, AU, BB, BZ, CA, CY, GB, GD, GH, GM, IE, IL, IN, KE, LK, LS, MN, MW, MZ, NZ, SD, SG, SL, SZ, TT, TZ, UG, ZA, ZW): **UNILEVER N.V. [NL/NL]; Weena 455, NL-3013 AL Rotterdam (NL)**.

(71) Applicant (for AG, AU, BB, BZ, CA, CY, GB, GD, GH, GM, IE, IL, KE, LK, LS, MN, MW, MZ, NZ, SD, SG, SL, SZ, TT, TZ, UG, ZA, ZW only): **UNILEVER PLC [GB/GB]; Unilever House, Blackfriars, London EC4P 4BQ (GB)**.

(71) Applicant (for IN only): **HINDUSTAN LEVER LTD [IN/IN]; Hindustan Lever House, 165-166 Backbay Reclamation, Mumbai 400 020 (IN)**.

(72) Inventors: **GOEDHART, Machiel; Unilever Research Vlaardingen, Olivier van Noortlaan 120, NL-3133 AT Vlaardingen (NL). KERPELS, Fred; Unilever Research Vlaardingen, Olivier van Noortlaan 120, NL-3133 AT Vlaardingen (NL). VAN KRALINGEN, Cornelis, Gerhard; Unilever Research Vlaardingen, Olivier**

van Noortlaan 120, NL-3133 AT Vlaardingen (NL). **OVERDEVEST, Pieter, E., M.; Unilever Research Vlaardingen, Olivier van Noortlaan 120, NL-3133 AT Vlaardingen (NL). VRIESWIJK, Karin; Unilever Research Vlaardingen, Olivier van Noortlaan 120, NL-3133 AT Vlaardingen (NL)**.

(74) Agents: **ROSEN JACOBSON, Frans, L., M. et al.; Patent Department, Olivier van Noortlaan 120, NL-3133 AT Vlaardingen (NL)**.

(81) Designated States (national): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PH, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, UZ, VN, YU, ZA, ZW.

(84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:

- with international search report
- before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.



A1  
WO 02/053824

(54) Title: DRY CLEANING COMPOSITIONS AND THEIR USE

(57) Abstract: A dry cleaning composition comprising: (a) more than 60 wt % to 99.9 wt % of an Isoparaffinic solvent having a molecular average of at least 9 carbon atoms; (b) from 0.01 wt % to 39 wt % of water; (c) from 0.01 wt % to 39 wt % of emulsifier; and (d) from 0.001 wt % to 39 wt % of a cleaning auxiliary.

**DRY CLEANING COMPOSITIONS AND THEIR USE****Technical Field**

The present invention relates to dry cleaning compositions  
5 based on organic solvents and processes.

**Background of the Invention**

Traditionally, dry cleaning processes are carried-out using organic solvents, in particular, perchloroethylene (PERC).

10 Stains are first removed by hand, typically using soap and/or surfactant, in a process known as "pre-spotting".

Recently, health and safety concerns surrounding PERC have lead to a search for alternative systems. For example, US-A-5 419 849 and US-A-5 454 969 disclose cleaning fluids for fabrics  
15 based on one or more hydrochlorofluorocarbons and/or hydrofluorocarbons, one or more alcohols and/or glycol ethers and one or more paraffinic hydrocarbons.

Another trend of recent times is to attempt to include in dry cleaning compositions, one or more auxiliary cleaning agents  
20 such as of the type used in conventional laundry wash products.

This has been achieved for dry cleaning systems which utilise liquid carbon dioxide instead of organic solvents. There, surfactant and water are used to create reverse micelles, e.g. as described in US-A-5 158 704 and US-A-5 266 205.

25 It is known to deliver hydrophilic fluorescers (a common laundry wash ingredient) in dry cleaning processes of the organic solvent type. US-A-3 640 881 describes how a hydrophilic fluorescer pre-mixed with water can be incorporated in a perchlorethylene (PERC) dry cleaning bath.

US-A-4 438 009 discloses a pre-spotting composition for use before dry cleaning, in which 1-30% of salts such as sodium citrate are included in a liquid comprising 5-60% of 5 hydrocarbon solvent such as limonene or an isoparaffin, 1-35% of nonionic surfactant and 40-75% of water.

We have now discovered that larger amounts of isoparaffinic solvents can be used to incorporate a wider range of cleaning auxiliaries and with greater efficacy.

10 Definition of the Invention

Thus, the present invention now provides dry cleaning composition comprising:

- (a) more than 60wt% to 99.9wt% of an isoparaffinic solvent having a molecular average of at least 9 carbon atoms, preferably from 70wt% to 99wt%, more preferably from 80wt% to 99wt%;
- (b) from 0.01wt% to 39wt% of water, preferably from 0.05wt% to 29wt%, more preferably from 0.1wt% to 19wt%;
- 20 (c) from 0.01wt% to 39wt% of emulsifier, preferably from 0.05wt% to 29wt%, more preferably from 0.1wt% to 19wt%; and
- (d) from 0.001wt% to 39wt% of a cleaning auxiliary, preferably from 0.01wt% to 29wt%, more preferably from 0.03wt% to 19wt%.

Detailed Description of the InventionThe Isoparaffinic Solvent

As used herein, the term "isoparaffinic solvent" means one or 5 more branched alkanes having on average, at least 9 carbon atoms, preferably from 10 to 16 carbon atoms.

Suitable isoparaffinic solvents, include those sold as DF-2000 or Isopar L, namely a mixture of C<sub>11</sub> to C<sub>15</sub> alkanes ex Exxon 10 Mobil. These isoparaffinic solvents are branched chain fully saturated hydrocarbons and are characterised by boiling range.

These mixtures are available in boiling ranges from 180°C to 210°C. In addition to the isoparaffinic hydrocarbons, low odor petroleum solvent having a boiling range of 195°C to 250°C, 15 kerosene and d-Limonene also are acceptable. From an odor standpoint, the isoparaffinic solvents are preferred, as these materials are low odor. However, if odor is not a consideration, substantially any of the above solvents can be utilised.

20

The Emulsifier

The compositions of the present invention also include an emulsifier, for example one or more surfactants selected from anionic, nonionic, cationic, zwitterionic and amphoteric 25 surfactants. One preferred selection comprises those disclosed in US-A-4 438 009. The first component of such a suitable nonionic surfactant mixture may be a sorbitan surfactant, such as sorbitan monolaurate, sorbitan mono-oleate, sorbitan

trioleate and mixtures thereof. The second component of the nonionic mixture includes the following classes of nonionic surfactants: the ethoxylated nonylphenols, such as the Surfonic N Series available from Jefferson Chemical the ethoxylated 5 octylphenols, including the Triton X Series available from Rohm & Haas, the ethoxylated secondary alcohols, such as<sup>3</sup> Tergitol Series available from Union Carbide, the ethoxylated primary alcohol series, such as the Neodols available from Shell Chemical, the polymeric ethylene oxides, such as the Pluronics 10 available from B.A.S.F. Wyandotte, and the ethylene oxide propylene oxide block copolymers, such as the Plurafacs available from B.A.S.F. Wyandotte.

The preferred nonionic surfactants recited in US-A-4 438 009 15 are the ethoxylated nonylphenols and the ethoxylated octylphenols, as these materials have excellent oil and water dispersibility, good detergency characteristics and can produce stable oil-out emulsions. The particularly preferred surfactants are nonylphenols having from 2 to 8 moles of 20 ethylene oxide, and particularly, nonylphenol having 6 moles of ethylene oxide combined with a small amount of nonylphenol reacted with 2 moles of ethylene oxide.

As an additional nonionic surfactant, it is often desirable to 25 incorporate an amount of an ethoxylated sorbitan nonionic, such as those sold under the tradename Tweens. Suitable nonionics include ethoxylated sorbitan monolaurate plus 20 moles ethylene oxide, ethoxylated sorbitan monopalmitate with 20 moles or less of ethylene oxide, ethoxylated sorbitan monostearate with 20 30 moles or less of ethylene oxide, ethoxylated sorbitan

monooleate with 20 moles or less of ethylene oxide and mixtures thereof. The Tween-type ethoxylated sorbitan nonionics, when combined with the non-ethoxylated sorbitan nonionics in appropriate amounts, provide excellent emulsion stability,  
5 increased stain removal performance and improved inhibition of soil redeposition.

In the most general sense, however, the emulsifier is chosen from one or more of soap and non-soap anionic, cationic, nonionic, amphoteric and zwitterionic surface-active compounds  
10 and mixtures thereof. Many suitable surface-active compounds are available and are fully described in the literature, for example, in "Surface-Active Agents and Detergents", Volumes I and II, by Schwartz, Perry and Berch.

15 Such surfactant(s) may be selected from one or more soaps and synthetic non-soap anionic and non-ionic compounds.

For example, the compositions of the invention may contain linear alkylbenzene sulphonate anionic surfactants,  
20 particularly linear alkylbenzene sulphonates having an alkyl chain length of C<sub>8</sub>-C<sub>15</sub>.

The compositions of the invention may additionally or alternatively contain one or more other anionic surfactants.  
25 Suitable anionic surfactants are well-known to those skilled in the art. These include primary and secondary alkyl sulphates, particularly C<sub>8</sub>-C<sub>15</sub> primary alkyl sulphates; alkyl ether sulphates; olefin sulphonates; alkyl xylene sulphonates; dialkyl sulphosuccinates; and fatty acid ester sulphonates.  
30 Sodium salts are generally preferred.

Nonionic surfactants that may be used include the primary and secondary alcohol ethoxylates, especially the C<sub>8</sub>-C<sub>20</sub> aliphatic alcohols ethoxylated with an average of from 1 to 20 moles of ethylene oxide per mole of alcohol, and more especially the C<sub>10</sub>-5 C<sub>15</sub> primary and secondary aliphatic alcohols ethoxylated with an average of from 1 to 10 moles of ethylene oxide per mole of alcohol. Non-ethoxylated nonionic surfactants include alkylpolyglycosides, glycerol monoethers, and polyhydroxyamides (glucamide).

10

It is often preferred if the level of total non-ionic surfactant is less than the total amount of anionic surfactant (if present).

15

Another class of suitable surfactants comprises certain mono-long chain-alkyl cationic surfactants. Cationic surfactants of this type include quaternary ammonium salts of the general formula R<sub>1</sub>R<sub>2</sub>R<sub>3</sub>R<sub>4</sub>N<sup>+</sup> X<sup>-</sup> wherein the R groups are long or short 20 hydrocarbon chains, typically alkyl, hydroxyalkyl or ethoxylated alkyl groups, and X is a counter-ion (for example, compounds in which R<sub>1</sub> is a C<sub>8</sub>-C<sub>22</sub> alkyl group, preferably a C<sub>8</sub>-C<sub>10</sub> or C<sub>12</sub>-C<sub>14</sub> alkyl group, R<sub>2</sub> is a methyl group, and R<sub>3</sub> and R<sub>4</sub>, which may be the same or different, are methyl or hydroxyethyl 25 groups); and cationic esters (for example, choline esters).

#### The Cleaning Auxiliary

Suitable cleaning auxiliaries may be selected from any one or 30 more of those included in conventional laundry wash compositions, e.g. selected from detergency builders, bleaches, alkaline agents, fluorescers and mixtures thereof.

Detergency Builders

Inorganic builders that may be present include sodium carbonate, if desired in combination with a crystallisation seed for calcium carbonate, as disclosed in GB 1 437 950 (Unilever); crystalline and amorphous aluminosilicates, for example, zeolites as disclosed in GB 1 473 201 (Henkel), amorphous aluminosilicates as disclosed in GB 1 473 202 (Henkel) and mixed crystalline/amorphous aluminosilicates as disclosed in GB 1 470 250 (Procter & Gamble); and layered silicates as disclosed in EP 164 514B (Hoechst). Inorganic phosphate builders, for example, sodium orthophosphate, pyrophosphate and tripolyphosphate are also suitable for use with this invention.

15

Another suitable class of builders comprises the alkali metal, preferably sodium, aluminosilicates.

20 The alkali metal aluminosilicate may be either crystalline or amorphous or mixtures thereof, having the general formula: 0.8-1.5 Na<sub>2</sub>O. Al<sub>2</sub>O<sub>3</sub>. 0.8-6 SiO<sub>2</sub>.

These materials contain some bound water and are required to 25 have a calcium ion exchange capacity of at least 50 mg CaO/g. The preferred sodium aluminosilicates contain 1.5-3.5 SiO<sub>2</sub> units (in the formula above). Both the amorphous and the crystalline materials can be prepared readily by reaction between sodium silicate and sodium aluminate, as amply described in the 30 literature. Suitable crystalline sodium aluminosilicate ion-exchange detergency builders are described, for example, in GB 1 429 143 (Procter & Gamble). The preferred sodium

aluminosilicates of this type are the well-known commercially available zeolites A and X, and mixtures thereof.

The zeolite may be the commercially available zeolite 4A now 5 widely used in laundry detergent powders. However, according to a preferred embodiment of the invention, the zeolite builder incorporated in the compositions of the invention is maximum aluminium zeolite P (zeolite MAP) as described and claimed in EP 384 070A (Unilever). Zeolite MAP is defined as an alkali metal 10 aluminosilicate of the zeolite P type having a silicon to aluminium ratio not exceeding 1.33, preferably within the range of from 0.90 to 1.33, and more preferably within the range of from 0.90 to 1.20.

15 Especially preferred is zeolite MAP having a silicon to aluminium ratio not exceeding 1.07, more preferably about 1.00. The calcium binding capacity of zeolite MAP is generally at least 150 mg CaO per g of anhydrous material.

20 Organic builders that may be present include polycarboxylate polymers such as polyacrylates, acrylic/maleic copolymers, and acrylic phosphinates; monomeric polycarboxylates such as citrates, gluconates, oxydisuccinates, glycerol mono-, di and trisuccinates, carboxymethyloxy succinates,

25 carboxymethyloxymalonates, dipicolinates, hydroxyethyliminodiacetates, alkyl- and alkenylmalonates and succinates; and sulphonated fatty acid salts. This list is not intended to be exhaustive.

30 Especially preferred organic builders are citrates and acrylic polymers, more especially acrylic/maleic copolymers.

Builders, both inorganic and organic, are preferably present in alkali metal salt, especially sodium salt, form.

Bleaches

5 Compositions according to the invention may also suitably contain a bleach system. Fabric washing compositions may desirably contain peroxy bleach compounds, for example, inorganic persalts or organic peroxyacids, capable of yielding hydrogen peroxide in aqueous solution.

10

Suitable peroxy bleach compounds include organic peroxides such as urea peroxide, and inorganic persalts such as the alkali metal perborates, percarbonates, perphosphates, persilicates and persulphates. Preferred inorganic persalts are sodium perborate 15 monohydrate and tetrahydrate, and sodium percarbonate.

Especially preferred is sodium percarbonate having a protective coating against destabilisation by moisture. Sodium percarbonate having a protective coating comprising sodium metaborate and 20 sodium silicate is disclosed in GB 2 123 044B (Kao).

The peroxy bleach compound is suitably present in an amount in excess of that of the bleach precursor.

25 Preferred bleach precursors are peroxycarboxylic acid precursors, more especially peracetic acid precursors and peroanoic acid precursors. Especially preferred bleach precursors suitable for use in the present invention are N,N,N',N',-tetracetyl ethylenediamine (TAED) and sodium 30 noanoyloxybenzene sulphonate (SNOBS). The novel quaternary ammonium and phosphonium bleach precursors disclosed in US 4 751 015 and US 4 818 426 (Lever Brothers Company) and EP 402 971A

(Unilever), and the cationic bleach precursors disclosed in EP 284 292A and EP 303 520A (Kao) are also of interest.

The bleach system can be either supplemented with or replaced by 5 a peroxyacid. Examples of such peracids can be found in US 4 686 063 and US 5 397 501 (Unilever). A preferred example is the imido peroxy carboxylic class of peracids described in EP A 325 288, EP A 349 940, DE 382 3172 and EP 325 289. A particularly preferred example is pthalimido peroxy caproic acid (PAP).

10

A bleach stabiliser (transition metal sequestrant) may also be present. Suitable bleach stabilisers include ethylenediamine tetra-acetate (EDTA), the polyphosphonates such as Dequest (Trade Mark) and non-phosphate stabilisers such as EDDS 15 (ethylene diamine di-succinic acid). These bleach stabilisers are also useful for stain removal especially in products containing low levels of bleaching species or no bleaching species.

20 An especially preferred bleach system comprises a peroxy bleach compound (preferably sodium percarbonate optionally together with a bleach activator), and a transition metal bleach catalyst as described and claimed in EP 458 397A, EP 458 398A and EP 509 787A (Unilever).

25

#### Enzymes

Compositions according to the invention may also contain one or more enzyme(s). Suitable enzymes include the proteases, amylases, cellulases, oxidases, peroxidases and lipases usable 30 for incorporation in detergent compositions. Preferred proteolytic enzymes (proteases) are, catalytically active protein materials which degrade or alter protein types of stains when present as in fabric stains in a hydrolysis

reaction. They may be of any suitable origin, such as vegetable, animal, bacterial or yeast origin.

Proteolytic enzymes or proteases of various qualities and 5 origins and having activity in various pH ranges of from 4-12 are available and can be used in the instant invention. Examples of suitable proteolytic enzymes are the subtilisins which are obtained from particular strains of B. Subtilis B. licheniformis, such as the commercially available subtilisins 10 Maxatase (Trade Mark), as supplied by Gist Brocades N.V., Delft, Holland, and Alcalase (Trade Mark), as supplied by Novo Industri A/S, Copenhagen, Denmark.

Particularly suitable is a protease obtained from a strain of 15 Bacillus having maximum activity throughout the pH range of 8-12, being commercially available, e.g. from Novo Industri A/S under the registered trade-names Esperase (Trade Mark) and Savinase (Trade-Mark). The preparation of these and analogous enzymes is described in GB 1 243 785. Other commercial proteases 20 are Kazusase (Trade Mark obtainable from Showa-Denko of Japan), Optimase (Trade Mark from Miles Kali-Chemie, Hannover, West Germany), and Superase (Trade Mark obtainable from Pfizer of U.S.A.).

## 25 Fluorescers

Fluorescers may also be included, either hydrophobic and/or hydrophobic types.

Some preferred classes of hydrophobic fluorescers are coumarins, eg Tinopal SWN and bis-benzoxazoles, eg. Tinopal SOP.

30 Some preferred classes of hydrophilic fluorescers are those which are organic fluorescers having one or more hydrophilic groups independently selected from sulphonate, carboxylate,

sulphate, phosphate, phosphonate, phosphinate, hydroxyl, (primary, secondary and tertiary) amino and (poly)alkoxylated groups. Some preferred types include water-soluble and water-dispersible distyrylbiphenyl derivatives, distilbene derivatives, especially cyanuric chloride/diaminostilbene derivatives and dibenzofuranbiphenyl derivatives.

Alkaline Agents

Suitable alkaline agents include alkali metal carbonates, silicates and metasilicates, alone or in combination.

10 The present invention will now be explained in more detail by way of the following non-limiting examples.

Example 1

Component	weight%	Detergent product	weight%
15 Isopar L	97	Na-citrate	25
Water	1.0	Sokalan CP45	3.3
Aerosol OT	0.67	TAED	14
Plurafac LF403	1.0	Na-percarbonate	41
20 Detergent product	0.11	Dequest 2047	2.0
		Light soda ash	8.4
		Na-CMC	1.1
		Tinopal CBS-X	3.4
		Savinase	2.1
25 Total	100	Total	100

Experimental procedure

This procedure describes the cleaning of oil (dirty motor oil, kitchen grease, spaghetti sauce), particulates (sebum, soot, clay) and bleachable (tea) stain monitors. In this example the 5 total monitor weight was 10.3 g and the liquid to cloth ratio was 13 g/g, requiring 134 g of emulsion fluid. Therefore, 0.90 g AOT and 1.34 g Plurafac LF403 were dissolved in 130 g DF-2000, and 0.15 g detergent product was dissolved in 1.34 g of water. Mixing both solutions together formed the emulsion. 10 The cleaning took place in a rotating closed container including both the emulsion and the stain monitors. Similarly, an identical set of monitors was washed with a traditional laundry detergent (5 g/L) and water.

15 Results

The emulsion showed excellent cleaning compared to cleaning with water and a traditional laundry detergent. Additionally, the emulsion resulted in very good fabric care compared to traditional water-based cleaning.

**Claims**

1. A dry cleaning composition comprising:
  - (a) more than 60wt% to 99.9wt% of an isoparaffinic solvent having a molecular average of at least 9 carbon atoms, preferably from 70wt% to 99wt%, more preferably from 80wt% to 98wt%;
  - (b) from 0.01wt% to 39wt% of water, preferably from 0.05wt% to 29wt%, more preferably from 0.1wt% to 19wt%;
  - (c) from 0.01wt% to 39wt% of emulsifier, preferably from 0.05wt% to 29wt%, more preferably from 0.1wt% to 19wt%; and
  - (d) from 0.001wt% to 39wt% of a cleaning auxiliary, preferably from 0.01wt% to 29wt%, more preferably from 0.03wt% to 19wt%.
2. A composition according to claim 1, when the isoparaffinic solvent has an average of from 10 to 16 carbon atoms, preferably from 11 to 15 carbon atoms.
3. A composition according to either preceding claim, wherein the emulsifier comprises one or more surfactants selected from ionic and nonionic surfactants and has an HLB of from 5 to 15.
4. A composition according to any preceding claim, wherein the weight ratio of water to emulsifier is from 20:1 to 1:20.
5. A composition according to any preceding claim, wherein the cleaning auxiliary is selected from detergency

15

builders, bleaches, alkaline agents, fluoresceors, enzymes and mixtures thereof.

6. A process for dry cleaning textile fabrics, the process comprising contacting said fabrics with a composition according to any preceding claim, at atmospheric pressure and at ambient temperature.

## INTERNATIONAL SEARCH REPORT

National Application No

PCT/EP 01/15048

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 D06L1/04

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 D06L C11D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the International search (name of data base and, where practical, search terms used)

WPI Data, EPO-Internal, PAJ

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4 013 575 A (CASTRANTAS HARRY MARCUS ET AL) 22 March 1977 (1977-03-22) claim 1; example 1 ---	1-6
Y	GB 2 238 793 A (KREUSSLER CHEM FAB) 12 June 1991 (1991-06-12) abstract; example 1 ---	1-6
A	US 5 419 849 A (FIELDS PAUL B ET AL) 30 May 1995 (1995-05-30) cited in the application example 2 ---	1,2,6
A	US 5 604 196 A (WELTMAN HENRY J ET AL) 18 February 1997 (1997-02-18) column 5, line 65 -column 6, line 6; table VII ---	1,2,6
		-/-



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

## \* Special categories of cited documents :

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
- \*E\* earlier document but published on or after the International filing date
- \*L\* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

- \*T\* later document published after the International filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- \*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- \*Y\* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- \*&\* document member of the same patent family

Date of the actual completion of the International search

6 May 2002

Date of mailing of the International search report

14/05/2002

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax (+31-70) 340-3016

Authorized officer

Saunders, T

## INTERNATIONAL SEARCH REPORT

National Application No

/EP 01/15048

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>DATABASE WPI Section Ch, Week 199512 Derwent Publications Ltd., London, GB; Class E19, AN 1995-085724 XP002169368 &amp; JP 07 011298 A (SANKEN KK), 13 January 1995 (1995-01-13) abstract</p> <p>-----</p>	1-3, 6

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 01/15048

Patent document cited in search report		Publication date		Patent family member(s)		Publication date
US 4013575	A	22-03-1977	NONE			
GB 2238793	A	12-06-1991	DE	3940804 A1		13-06-1991
US 5419849	A	30-05-1995	US	5454969 A		03-10-1995
US 5604196	A	18-02-1997	US	5437808 A		01-08-1995
			US	5188754 A		23-02-1993
			CA	2141433 A1		17-02-1994
			EP	0672101 A1		20-09-1995
			IL	106551 A		22-09-1999
			JP	2791217 B2		27-08-1998
			JP	7508790 T		28-09-1995
			KR	128262 B1		03-04-1998
			MX	9304742 A1		30-06-1994
			WO	9403579 A1		17-02-1994
			MX	9206460 A1		28-02-1994
			DE	69230176 D1		25-11-1999
			EP	0598023 A1		25-05-1994
			US	5370817 A		06-12-1994
			WO	9303102 A1		18-02-1993
			US	5416253 A		16-05-1995
JP 7011298	A	13-01-1995	JP	2517524 B2		24-07-1996

THIS PAGE BLANK (USPTO)